

**TRANSMITTAL LETTER TO THE UNITED STATES  
DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING  
A FILING UNDER 35 U.S.C. 371**

U.S. APPLICATION NO. (if known, see 37 CFR 1.5)

**09/555899**INTERNATIONAL APPLICATION NO.  
PCT/EP98/07868INTERNATIONAL FILING DATE  
December 3, 1998PRIORITY DATE CLAIMED  
December 4, 1997TITLE OF INVENTION  
APPARATUS FOR SELECTIVE CATALYTIC OXIDATION OF CARBON MONOXIDEAPPLICANT(S) FOR DO/EO/US  
Stefan BRAUCHEL, Oliver FREITAG, Barbara STROBEL and Matthias WOLFSTEINER

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1.  This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2.  This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3.  This express request to begin national examination procedures (35 U.S.C. 371(f) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1)).
4.  A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5.  A copy of the International Application as filed (35 U.S.C. 371(c)(2)).
  - a.  is transmitted herewith (required only if not transmitted by the International Bureau).
  - b.  has been transmitted by the International Bureau
  - c.  is not required, as the application was filed in the United States Receiving Office (RO/US)
6.  A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7.  Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
  - a.  are transmitted herewith (required only if not transmitted by the International Bureau).
  - b.  have been transmitted by the International Bureau.
  - c.  have not been made; however, the time limit for making such amendments has NOT expired.
  - d.  have not been made and will not be made.
8.  A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9.  An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). (UNEXECUTED)
10.  A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

**Item 11. to 16. below concern other document(s) or information included:**

11.  An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12.  An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13.  A **FIRST** preliminary amendment.
14.  A **SECOND** or **SUBSEQUENT** preliminary amendment.
15.  A substitute specification.
16.  A change of power of attorney and/or address letter.
16.  Other items or information:  
PCT/IB308

US APPLICATION NO. 09/555899		INTERNATIONAL APPLICATION NO PCT/EP98/07868	ATTORNEY'S DOCKET NUMBER 225/48888
17. <input checked="" type="checkbox"/> The following fees are submitted:		CALCULATIONS PTO USE ONLY	
Basic National Fee (37 CFR 1.492(a)(1)-(5)): Search Report has been prepared by the EPO or JPO .....		\$ 840.00	
International preliminary examination fee paid to USPTO (37 CFR 1.482) ...		\$ 670.00	
No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)) .....		\$ 760.00	
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO .....		\$ 970.00	
International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 53(2)(4) .....		\$ 96.00	
<b>ENTER APPROPRIATE BASIC FEE AMOUNT</b>		\$ 840.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input checked="" type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).		\$ 130.00	
Claims	Number Filed	Number Extra	Rate
Total Claims	13 - 20=	0	X \$18.00
Independent Claims	1 - 3=	0	X \$78.00
Multiple dependent claims(s) (if applicable)		+ \$260.00	
<b>TOTAL OF ABOVE CALCULATIONS</b>		\$ 970.00	
Reduction by 1/2 for filing by small entity, if applicable. Verified Small Entity statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28).		\$	
<b>SUBTOTAL</b>		\$	
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input checked="" type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).		\$	
<b>TOTAL NATIONAL FEE</b>		\$	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28.3.1). \$40.00 per property +		\$	
<b>TOTAL FEE ENCLOSED</b>		\$ 970.00	
		Amount to be: refunded	\$
		charged	\$
<p>a. <input checked="" type="checkbox"/> One check in the amount of \$ 970.00 for the filing fee is enclosed</p> <p>b. <input type="checkbox"/> Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed.</p> <p>c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees, which may be required, or credit any overpayment to Deposit Account No. <u>05-1323</u>. A duplicate copy of this sheet is enclosed.</p>			
<p>NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.</p> <p><i>Wa A 20 13085</i></p>			
<p>SEND ALL CORRESPONDENCE TO:</p> <p>Evenson, McKeown, Edwards &amp; Lenahan, P.L.L.C. 1200 G Street, N.W., Suite 700 Washington, D.C. 20005 Tel. No. (202) 628-8800 Fax No. (202) 628-8844</p>			
<p><i>fw</i></p> <p>SIGNATURE Donald D. Evenson NAME 26,160 REGISTRATION NUMBER June 5, 2000 DATE</p>			

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: STEFAN BRAUCHLE ET AL.

Int'l App. No.: PCT/EP98/07868

Serial No.: Group Art Unit:

Filed: June 5, 2000 Examiner:

Title: DEVICE FOR SELECTIVE CATALYTIC OXIDATION  
OF CARBON MONOXIDE

PRELIMINARY AMENDMENT

Box PCT  
Commissioner for Patents  
Washington, D.C. 20231

Sir:

Please enter the following amendments to the specification and claims, as amended by way of Annexes to the International Preliminary Examination Report for PCT/EP98/07868, prior to the examination of the application during the U.S. National Phase.

IN THE SPECIFICATION:

Page 1, line 8, add --BACKGROUND AND SUMMARY OF THE INVENTION--.

Page 1, line 10-11, delete "according to the precharacterizing clause of Patent Claim 1".

Page 1, lines 36-37, delete "having the features of Patent Claim 1" and insert therefor --according to the present invention--.

Page 2, line 22, add --BRIEF DESCRIPTION OF THE DRAWINGS--.

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Page 3, line 12, add --DETAILED DESCRIPTION OF THE DRAWINGS--.

Page 3, line 24, change "reformat" to --reformat--.

Page 13, line 4, change "reformat" to --reformat--.

Page 16, line 5, change "Patent Claims" to --WHAT IS CLAIMED IS--.

IN THE CLAIMS:

Please cancel Claims 1-12, without prejudice to or disclaimer of the subject matter therein.

Please add new Claims 13-25 as follows:

--13. A multistage apparatus for selective catalytic oxidation of carbon monoxide contained in a hydrogen-rich mixed gas flow, comprising:

a plurality of reaction chambers filled with a catalyst and a plurality of cooling chambers through which a coolant flows, said reaction chambers and coolant chambers being formed between successive plates and defining at least two stages in a plate stack,

wherein said plates have openings that form inlet or outlet channels for the reaction chambers and for the cooling chambers,

wherein the reaction chambers are each connected exclusively to inlet and outlet channels for the mixed gas flow,

wherein the cooling chambers are each connected exclusively to inlet and outlet channels for the coolant;

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a separating plate without any opening for inlet channels for the mixed gas flow and arranged between at least two successive stages; and

an first apparatus for supplying an oxidizing gas to a first stage;

an second apparatus for supplying an oxidizing gas to at least one other stage following the first stage, wherein the second apparatus leads into an outlet channel for the mixed gas flow from a preceding stage.

14. An apparatus according to Claim 13, comprising three stages and further comprising:

an additional separating plate without any opening for the inlet channels for the mixed gas flow for a second stage and arranged between the second and a third stage; and

an third apparatus for supplying an oxidizing gas to the third stage leading into an outlet channel for the mixed gas flow from the second stage.

15. An apparatus according to Claim 13, wherein the second apparatus for supplying the oxidizing gas is a tubular probe that passes through an end plate into an outlet channel from the first stage.

16. An apparatus according to Claim 13, wherein the second apparatus for supplying the oxidizing gas is a tubular probe that passes through an end plate and an inlet channel for a second stage.

17. An apparatus according to Claim 14, wherein the second apparatus for supplying the oxidizing gas is a tubular probe that passes through an end plate or passes through the

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end plate, the outlet channel from the third stage, the separating plate between the second and the third stage, an inlet channel of the second stage, and into an outlet channel from the first stage.

18. An apparatus according to Claim 14, wherein the third apparatus for supplying the oxidizing gas is a tubular probe that passes through an end plate and, in an opposite direction to the mixed gas flow, through an inlet channel of the third stage, into an outlet channel from the second stage.

19. An apparatus according to Claim 14, wherein the third apparatus for supplying the oxidizing gas is a tubular probe that passes through the supply channel of the first stage and through a separating plate between the first and the second stage, into the outlet channel from the second stage.

20. An apparatus according to Claim 13, wherein the first or second apparatus for supplying the oxidizing gas is a tubular probe having a plurality of outlet openings arranged at points where individual reaction chambers enter the outlet channels for the mixed gas flow, in each case corresponding to a level of a reformat flow.

21. An apparatus according to Claim 20, wherein the plurality of outlet openings are arranged radially around the probe.

22. An apparatus according to Claim 20, wherein the plurality of outlet openings face the outlet channels.

23. An apparatus according to Claim 13, wherein the second apparatus for supplying the oxidizing gas is a tubular, coaxial probe having an inner tube which, at one end has outlet openings into an outer tube and wherein the outer tube, on an opposite end, has outlet openings into an inlet channel of a stage.

24. An apparatus according to Claim 13, wherein the second apparatus for supplying the oxidizing gas is a tubular probe comprising at one end an inner tube having one or more outlet openings into a deflection device and, on an opposite end has one or more outlet openings into an outlet channel from the preceding stage.

25. An apparatus according to Claim 13, wherein the oxidizing gas from the second apparatus is supplied into an area between an outlet channel of the preceding stage and an inlet channel of the following stage, in which area gas flows from the reaction chambers are combined to form an overall volume flow.--

**IN THE ABSTRACT:**

Please substitute the new Abstract of the Disclosure submitted herewith on a separate page for the original Abstract presently in the application.

**REMARKS**

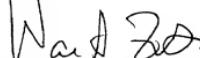
Entry of the amendments to the specification, claims, and abstract, as amended by way of Annexes to the International Preliminary Examination Report for PCT/EP98/07868, before examination of the application in the U.S. National Phase is respectfully requested.

If there are any questions regarding this Preliminary Amendment or this application in general, a telephone call to the undersigned would be appreciated since this should expedite the prosecution of the application for all concerned.

If necessary to effect a timely response, this paper should be considered as a petition for an Extension of Time sufficient to effect a timely response, and please charge any deficiency in fees or credit any overpayments to Deposit Account No. 05-1323 (Docket #225/48888).

June 5, 2000

Respectfully submitted,

  
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--ABSTRACT

An apparatus for the selective catalytic oxidation of carbon monoxide contained in a hydrogen-rich mixed gas flow, includes a reaction chamber filled with a catalyst and a cooling chamber through which a coolant flows formed between successive plates. The plates have openings that form inlet and outlet channels for the mixed gas flow and the coolant. An oxidizing gas is supplied directly into the inlet channel for the mixed gas flow. For two-stage apparatuses, a separating plate without an opening for the inlet channel for the mixed gas flow is arranged between the first and the second stage, and an apparatus for supplying the oxidizing gas to the second stage is passed into the outlet channel for the mixed gas flow from the first stage.--

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Apparatus for selective catalytic oxidation of carbon monoxide

10 The invention relates to an apparatus for selective catalytic oxidation of carbon monoxide in a hydrogen-rich mixed gas flow according to the precharacterizing clause of Patent Claim 1.

15 An apparatus of this generic type and having a plate-type construction, which is intended to reduce the proportion of carbon monoxide in the gas produced by a water-vapour reformer, is disclosed in DE 195 44 895 Cl. In this arrangement, plates with reaction chambers and heat-exchanger chambers are stacked on one another alternately. Each plate has four through-openings in each 20 case for the mixed gas flow or heat carrier medium inlet and outlet. In order to allow air to be supplied with a different volume flow in each stage, holes can also be provided on the end faces of the plates having the reaction chambers, through each of which holes air can be 25 metered into the reaction chambers. However, additional components need to be provided in order to distribute the air.

Furthermore, DE 87 09 386 U1 discloses an apparatus for using a probe, arranged in the flow 30 direction of the first gas, in order to apply a second gas to a reaction chamber through which gas flows.

The object of the invention is to provide an apparatus for selective catalytic oxidation of carbon monoxide in a hydrogen-rich mixed gas flow, with 35 simplified supply and mixing for the oxidizing gas.

This object is achieved by an apparatus having the features of Patent Claim 1.

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Integration of the supply for the oxidizing gas into the inlet channel for the mixed gas flow represents a solution that is simple and cost-effective for production. In a first advantageous configuration, the 5 supply ends in the manifold channel, where there is no catalyst material. There is thus no need for any additional mixing path or distributor structures. Furthermore, the ratio of the reaction and cooling 10 chambers to one another can be varied as required, in terms of volume and/or the number of plates.

A plurality of stages can be integrated in one physical unit, without any problems. In this case, high 15 flexibility is nevertheless maintained by the variable inlet position for supplying the oxidizing gas. Overall, the apparatus according to the invention has improved 20 characteristics in terms of volume requirement, weight and costs.

Advantageous configurations of the apparatus for supplying the oxidizing gas results in reliable 25 homogenization of the gas mixture between each individual stage.

Further advantages and refinements of the invention result from the dependent claims and the description. The invention is described in more detail in 30 the following text with reference to a drawing, in which: Figure 1 shows an exploded illustration of the basic construction of a plate apparatus for selective catalytic oxidation of carbon monoxide in a hydrogen-rich mixed gas flow, Figure 2 shows a section through the apparatus as shown in Figure 1, along the line A-A, for a two-stage design, Figure 3 shows a section through the apparatus as shown in Figure 1, along the line B-B, likewise for 35 a two-stage design, Figure 4 shows a section through the apparatus as shown in Figure 1, along the line A-A, for a three-

stage design, and

Figure 5 shows a section through the apparatus as shown in Figure 1, along the line B-B, likewise for a three-stage design,

5 Figure 6 shows a section through the apparatus as shown in Figure 1, along the line A-A, for a three-stage design having a probe,

Figure 7 shows an advantageous refinement of a probe as shown in Figure 4, and

10 Figure 8 shows a further advantageous refinement of a probe.

The apparatus which is denoted overall by 1 in Figure 1, but only part of which is shown, for selective catalytic oxidation of carbon monoxide contained in a hydrogen-rich mixed gas flow comprises a large number of plates 2 stacked on one another, with reaction chambers 4 and cooling chambers 5 being formed between adjacent plates 2 after assembly. For simplicity, Figure 1 shows only some of the plates 2. The overall design of the 15 apparatus 1 can be seen in Figures 2-7.

The apparatus shown in the exemplary embodiment for selective catalytic oxidation, referred to as a CO oxidizer 1 in the following text, is preferably used to remove carbon monoxide from the reformat from an 20 apparatus for water-vapour reformation of a crude fuel, for example methanol. Such reformers are used in particular to produce hydrogen for mobile fuel cell applications. Fuel cells having an electrolyte membrane which passes protons, so-called PEM fuel cells, in 25 particular, react to carbon monoxide contained in the hydrogen gas by producing toxins. The aim is thus to reduce the proportion of carbon monoxide in the hydrogen gas, using a CO oxidizer 1, by selective catalytic oxidation of the carbon monoxide with the addition of oxygen, on a suitable oxidation catalyst. Multistage CO 30 oxidizers 1 are preferably used to reduce the proportion of carbon monoxide to values less than 50 ppm, with

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oxygen being supplied separately to each stage.

Although the invention is described in the following text on the basis of a CO oxidizer 1 for removal of carbon monoxide from the gas produced by an 5 apparatus for water-vapour reformation from methanol, the field claimed is not intended to be limited to this application.

The oxygen is preferably supplied in the form of a predetermined amount of oxygen in the air. Metering 10 apparatuses, which are not shown, can be provided for this purpose. Suitable catalyst materials for selective catalytic CO oxidation include platinum and/or ruthenium on a zeolite or aluminium oxide base. This catalyst material may be introduced into the reaction chamber 4 in 15 any desired form. Apart from using granular material or pellets, it is also possible to apply the catalyst material to the plate surfaces or to the catalyst support units.

The addition of oxygen leads to the following 20 competing oxidation reactions in the mixed gas flow containing hydrogen and carbon monoxide:



and



The catalyst material and the operating 30 conditions are chosen in such a manner that the first reaction takes place in preference to the second reaction, that is to say selectively, and thus, although the proportion of CO in the mixed gas flow is reduced, as little hydrogen as possible is consumed at the same time. Since both reactions are exothermal, it is necessary to remove the heat produced, for example using a coolant 35 from the CO oxidizer 1. Cooling chambers 5 are provided in the CO oxidizer 1 for this purpose.

Projections 6 are provided along each edge of the

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plates 2, and each come into contact with the underneath of the adjacent plate 2 during assembly, with the projections 6 and the underneath of the adjacent plate 2 being connected to one another in a gastight manner by 5 using seals, or preferably by welding. This results in the reaction and cooling chambers 4, 5, which have already been described further above. The plates 2 also each have openings 3 - referred to as holes in the following text - for the input and output, respectively, 10 of the mixed gas flow and of the cooling media, respectively. Furthermore, annular projections 7 are arranged in places around the holes 3, and are likewise connected in a gastight manner to the underneath of the respective adjacent plate 2 during assembly of the CO oxidizer 1. Further flow guidance structures can be provided in the reaction chambers and cooling chambers 15 4, 5, but these are not shown in the drawing, for simplicity.

After assembly, the holes 3 in the individual 20 plates 2 overall form an inlet and outlet channel 11, 12, respectively, for the mixed gas flow. The holes 3 form corresponding inlet and outlet channels 13, 14, respectively, for the coolant. The direction in which the media are carried in the channels 11-14 and the 25 corresponding reaction chambers and cooling chambers 4, 5, respectively, are represented by dotted arrows in the drawing.

In order to form the reaction chambers 4 and 30 cooling chambers 5, all the holes 3 through which the mixed gas flow or the coolant flows are now surrounded by an annular projection 7 for each plate 2. The reaction chambers 4 are thus connected, for flow purposes, only to the inlet and outlet channel 11, 12, respectively, for the mixed gas flow, while the cooling chambers 5 are 35 connected, for flow purposes, only to the inlet or outlet channel 13, 14, respectively, for the coolant. This prevents the mixed gas flow from being mixed with the

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coolant.

In the illustrated exemplary embodiment, the appropriate medium in each case flows through the reaction and cooling chambers 4, 5, respectively, 5 transversely and in the opposite direction to the flow. However, the arrangement of the channels 11-14 and the flow directions can be matched to the requirement in virtually any desired manner. Even a cross-flow arrangement is possible. Furthermore, the embodiment 10 shown in Figure 1 corresponds to an apparatus having at least two stages, in which the oxygen required in the first stage I is supplied to the mixed gas flow upstream of the first stage I, via an external input line 10. However, it is also possible to supply the oxygen 15 directly into the inlet channel 11 of the first stage.

The oxygen for the second stage II is preferably introduced into the outlet channel 12a for the mixed gas flow from the first stage I using a probe 18. In the simplest case, the probe 18 is in the form of a tubular 20 pipe with any desired cross section. Since the oxygen is introduced into the outlet channel 12a from the first stage I, it is not available for the reaction in this first stage. In fact, the oxygen can mix, within the outlet channel 12a, with the mixed gas flowing out of the 25 first stage I. Since the outlet channel 12a from the first stage I is also at the same time used as the inlet channel 12b for the second stage II, a homogenous gas/oxygen mixture is thus supplied to the reaction chambers 4 in the second stage II. Additional external 30 mixing or distribution structures can thus be dispensed with. Further advantageous refinements of a probe 18 and of an oxygen supply are illustrated in Figures 6 and 7.

The CO oxidizer 1 illustrated in Figures 2 and 3 comprises two oxidation stages, referred to as I and II. 35 Figures 2 and 3 show section illustrations through the CO oxidizer 1, part of which is shown in Figure 1, in the area of the inlet and outlet channels 11-14 for the mixed

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gas flow (2), and for the coolant (Figure 3). A separating plate 16 is arranged in the plate stack, in order to form two stages I, II. No hole 3 for the inlet channel 11a is provided in the separating plate 16. The 5 inlet channel 11a for the mixed gas flow is thus sealed by the separating plate 16 at the end of the first stage I. The mixed gas flow is thus distributed between the reaction chambers 4 in the first stage I, and is collected again in the outlet channel 12a, after flowing 10 through the reaction chambers 4. In the exemplary embodiment, only two reaction chambers 4 are provided in each of the two stages I, II. However, the number may be varied as required, thus achieving good scaleability for the overall CO oxidizer 1, and for the individual stages 15 I, II as well. The oxygen required in the first stage I is added to the mixed gas flow, via a supply line 10, even before the first stage. However, it is also possible to supply the oxygen directly to the first stage I, via a further probe.

20 The function of the channels 11, 12 for the mixed  
gas flow is interchanged in the second stage II. The  
outlet channel 12a from the first stage I becomes the  
inlet channel 12b in the second stage II. In a  
corresponding manner, the outlet channel 11b in the  
25 second stage II is arranged at the point provided for the  
inlet channel 11a in the first stage I. The flow  
direction for the mixed gas flow in the second stage II  
is thus the opposite of that in the first stage I. The  
flow direction in the cooling chambers 5 remains the  
30 same.

The plate stack is bounded by end plates 15, 17, in which only some of the normal holes 3 are provided. The inlet channel 12b for the mixed gas flow is also sealed at the end of the second stage II by the upper end plate 17. The mixed gas flow is once again distributed between the reaction chambers 4 in the second stage II and is collected in the outlet channel 11b, after flowing

through the reaction chambers 4, and is carried out of the CO oxidizer 1. In a corresponding manner, only one hole 3 for the inlet channel or outlet channel 13, 14 is in each case arranged in the end plates 17, 15, 5 respectively, on the coolant side.

The oxygen for the second stage II is introduced into the outlet channel 12a from the first stage I using a probe 18. In the simplest case, the probe 18 is in the form of a single tube with a circular cross section. 10 However, apart from a tubular probe 18, any other desired cross-sectional shapes may also be used. Figures 6 and 7 show preferred configurations. Since the oxygen is introduced into the outlet channel 12a from the first stage I, it is not available for the reaction in this 15 first stage I. In order to distribute the oxygen better, the probe 18 may also be provided with radial outlet openings, in which case the axial opening of the probe 18 may be sealed. If the probe 18 is arranged in such a manner that the oxygen emerges in the flow direction of 20 the mixed gas flow, then radial deflection apparatuses, which cause the oxygen to mix better with the mixed gas flow, can also be provided.

According to the exemplary embodiment, the probe 18 is passed directly from underneath, through the end 25 plate 15, into the outlet channel 12a from the first stage I. However, it is also possible for the probe 18 to be passed from above, through the end plate 17 and the inlet channel 12b of the second stage II, into the outlet channel 12a from the first stage I.

30 The use of such a probe 18 has the advantage that, if the stack thickness varies, the point at which the oxygen enters the outlet-channel 12a can easily be adapted by changing the length of the probe 18. Furthermore, there is no need to provide holes or the 35 like in each plate in order to distribute and meter the oxygen adequately, leading to the apparatus being simplified, and thus to a reduction in costs.

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The concept described above can also be applied to apparatuses having three or more stages. For example, Figures 4 and 5 show a three-stage CO oxidizer 1, in which identical parts are denoted by the same reference symbols as in the other figures. However, in contrast to Figures 2 and 3, a further separating plate 16 is provided between the second and the third stage II, III. The invention is also suitable for apparatuses having four or more stages, in which the oxidizing medium is accordingly supplied, in a corresponding manner, between two successive stages.

The flow routing in Figure 4 corresponds to the exemplary embodiment shown in Figure 2, as far as and including the second stage II. The outlet channel 11b for the mixed gas flow from the second stage II is, however, at the same time used as the inlet channel 11c for the third stage III in this case. However, in addition, a second probe 20 for supplying oxygen in the opposite flow direction to that of the mixed gas flow is also introduced from above through the separating plate 17 into this inlet channel 11c, extends into the area of the outlet channel 11b from the second stage II, and ends there. The additional oxygen is thus introduced into the gas produced from the second stage, and can mix there with the mixed gas flow. Thus, once again, a homogeneous gas/oxygen mixture is supplied to the third stage III. The design options explained further above with reference to the probe 18 also apply to the probe 20, of course.

Since the inlet channel 11c ends before the end plate 17, the mixed gas flow is once again forced to flow the reaction chambers 4 in the third stage III and cannot leave the CO oxidizer 1, through the outlet channel 12c from the third stage III, until it has done so. As has already been described above, even with multistage apparatuses, there are no problems in varying the number and the sequence of the reaction chambers and cooling chambers 4, 5, respectively, in the individual stages.

Simple scaleability can thus also be ensured in multistage arrangements.

However, contrary to the situation shown in Figures 4 and 5, the probe 20 can also be passed from 5 underneath through the inlet channel 11a for the mixed gas flow for the first stage I, and through the separating plate 16 between the first and the second stage I, II, into the outlet channel 11b from the second stage II. In principle, in all the arrangements, it is 10 possible to introduce the probes 18, 20 into the respective outlet channel at the side, for example. However, in terms of production of the CO oxidizer, this is more complex and thus makes little sense.

No separating plates 16 are provided for the 15 coolant side in the exemplary embodiments. However, in this case as well, it is also possible to provide such separating plates 16, so that two or more sets of cooling chambers are produced, arranged in series. Furthermore, contrary to the exemplary embodiment, the cooling media 20 can also flow in the same direction as the mixed gas flow. In addition, an endothermic reaction can be caused to occur in the cooling chambers 5, in order to cool the CO oxidizer 1. In the case of gas generating systems for fuel cells, water and methanol, for example, could be 25 vaporized in the cooling chambers 5, thus extracting heat from the reaction chambers 5. In this context, the coolant would be the liquid to be vaporized. At the same time, it is possible to cause a reformation reaction to take place in the cooling chambers. To achieve this, the 30 cooling chambers 5 are filled with a suitable catalyst. A water vapour/methanol mixture is then passed as the coolant through the cooling chambers 5, with heat once again being extracted from the reaction chambers 4 of the CO oxidizer 1 during the endothermic reformation process.

35 A further advantageous refinement of an oxygen supply is illustrated in Figure 6. In principle, the tubular probes described in the exemplary embodiments may

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have any desired cross section, such as a circular, oval or polygonal cross section. The arrangement of the reaction and cooling chambers 4, 5 is comparable to the three-stage apparatus illustrated in the previous 5 figures. Identical elements are provided with the same reference symbols. A probe 21 extends through a stage  $n+1$  to the stage  $n$ . The probe is sealed at its lower end 21.3.

The probe 21 is provided along its longitudinal 10 axis with small holes, two of which 21.1, 21.2 are shown, which are arranged at the points where the individual channels 4 each enter the inlet channel 11b, corresponding to the level of the reformat flow that is flowing in. The holes 21.1, 21.2 can be arranged on one 15 side of the probe 21, preferably in the inlet flow side of the reformat, and/or may also each be distributed radially around the probe 21. The oxygen or air volume flow is thus supplied into the manifold channel 11b to correspond to the reformat volume flow that exists 20 locally, so that a suitable amount of oxygen is in each case present locally. On entering the inlet channel 11c, the media are then homogeneously mixed. If the metering were carried out too early, an explosive mixture could otherwise be produced, and if metering were carried out 25 too late, uniform media distribution problems could easily occur. This can be avoided by this refinement. Frits or other conventional metering devices are also suitable for use as the probe for metering.

The solution allows the reactor to have a compact 30 construction, while allowing a plurality of reactor stages to be integrated in one physical unit. Integration in the manifold or outlet channel avoids the need for any mixing part, which is also advantageous for physically separate reactors.

35 The ratio of the various reaction chambers, in particular the volume or plate thickness, can be varied as required, and the oxygen metering can easily be

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adapted during production by varying the distance between the holes 21.1, 21.2 in the longitudinal direction of the probe 21, and/or the diameter of the holes 21.1, 21.2, in a corresponding manner.

5 It is also possible to provide local metering of  
the oxygen only at every second, third or n-th inlet  
point, rather than at each point where the reformat  
enters the outlet channel 11. In the illustrated example,  
oxygen would then be metered only into the reformat from  
10 one channel, rather than into the reformat in both  
channels 4. Manufacture of appropriate probes 21 for  
arrangements with more than two cooling and/or reaction  
chambers per reactor stage can thus be simplified, with  
the costs being reduced.

15 A further advantageous option, in particular in  
a relatively long outlet channel having a relatively  
large number of reaction chambers, is to provide the  
holes in the probe 21 with an offset. This means that,  
for example, oxygen is metered only from a further  
20 reaction chamber in the outlet channel, for example the  
third or fourth, rather than at the first, foremost  
reaction chamber in the outlet channel.

Figure 7 shows a detailed illustration of a further advantageous embodiment of a probe 22, which likewise allows the media to be mixed reliably and well. The fitting of conventional means to generate turbulence in order to mix the media better has little advantage owing to the formation of undesirable pressure gradients in the inlet and/or outlet channel.

30 One advantage arrangement is for the probe 22 to be formed with double walls, with the oxygen being supplied through an inner tube 22.1, while the reformat is carried from the outlet channel 12 from the preceding stage in the tube 22.2, which surrounds the inner tube 22.1. The outer tube 22.2 may be arranged coaxially around the inner tube 22.1, or else eccentrically with respect to the inner tube 22.1. It is particularly

expedient for the oxygen outlet to be provided through holes 23 in the inner tube 22.1 in the area of the outlet channel, in which area the entire volume flow of the reformat from the preceding stage is joined together, 5 that is to say roughly at the level between the last reaction chamber in the preceding stage  $n$  and the first reaction chamber in the next stage  $n+1$ . In the example in Figure 2, the corresponding stages are  $n=1$  and  $n+1=2$ .

10 The design of the probe is particularly suitable for systems with a wide load spread, for which a wide dynamic range is required.

15 The medium preferably flows in the opposite direction in the inner tube 22.1 to that in the outer tube 22.2. At the lower end, the oxygen passes through holes 23 out of the inner tube 22.1 into the outer tube 22.2, where it joins the reformat flowing in from the preceding stage. In the outer tube 22.2, the mixture is passed to holes 24 at the upper end of the outer tube 22.2, flowing in the opposite direction to the flow in 20 the inner tube 22.1, and is mixed along the flow path. The mixture, which is now homogeneous, passes through the holes 24 in the outer tube 22.2 into the inlet channel of the reaction chambers in the next stage  $n+1$ , and at least part of it flows in the direction opposite to the flow 25 direction in the outer tube 22.2, into the inlet channel of the reactor stage.

30 Additional mixing elements, which are not illustrated, and turbulence generators can now also be arranged between the outer tube 22.2 and the inner tube 22.1 in order to mix the media better, without this causing any pressure gradient in the inlet channel to the reaction chambers in the next stage, since the mixing region is separate from the inlet channel 12b. In Figure 7, the probe is introduced into the inlet channel 35 or outlet channel in the opposite direction to the flow, although it is also possible to introduce it in the flow direction.

Simple hydrodynamic measures may be adopted in order to keep any pressure loss in the inlet channel 12b as small as possible. One advantageous measure is to reduce the diameter of the coaxial probe 22 and/or to 5 enlarge the cross section of the inlet channel 12.

Figure 8 shows a further advantageous configuration of a probe for supplying oxygen or air. This figure illustrates a probe 25 with an inner tube 25.1 and a deflection device 25.2 in the channel 12, in 10 which oxygen is introduced into the channel 12, in the direction of the flow in the inner tube 25.1. The oxygen emerges from the inner tube 25, preferably at the lower, open end or through radial openings in the tube wall, and is deflected in the deflection device 25.2 so that the 15 oxygen flows counter to the flow of the gas in the channel 12, between the tube 25.1 and the outer boundary of the deflection device 25.2. The oxygen in the deflection apparatus 25.2 is preferably passed through an opening 25.3 into the area of the channel 12a, in which 20 area the individual gas flows from the reaction chambers in the preceding stage n are joined together to form the overall volume flow, and pass into the area of the inlet channel for the following stage n+1, so that the oxygen is mixed with the overall volume flow before the mixed 25 gas flow in the inlet channel 12b is split between the reaction chambers 4 in the following stage n+1.

The reversal apparatus 25.2 is preferably a piece of tubing which is closed (25.4) at one end in the inner tube in the flow direction of the oxygen and essentially 30 coaxially surrounds the inner tube 25.1, or some comparable arrangement.

Various different embodiments of the probes (18, 20, 21, 22, 25) may also be used in combination in a single reactor.

35 It is also possible for the oxidizing medium to be metered in the input area upstream of the first stage of the reactor. This is advantageous if the reactor is

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preceded by another reactor, for example a reformer having a plate-type construction, or a heat exchanger. The probe for supplying the oxidizing medium is preferably introduced into the outlet flow from the upstream reactor.

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Patent Claims

1. Multistage apparatus (1) for selective catalytic oxidation of carbon monoxide contained in a hydrogen-rich mixed gas flow and having a plate-type construction,  
10 wherein

- all the stages are contained in a common plate stack,
- a reaction chamber (4) filled with a catalyst or a cooling chamber (5) through which a coolant flows  
15 is in each case formed between successive plates (2),
- plates (2) have openings (3) in order to form inlet or outlet channels (11-14), respectively, for the reaction chambers and cooling chambers (4, 5), respectively, and the reaction chambers and cooling chambers (4, 5), respectively, are each connected, for flow purposes, exclusively to the inlet and outlet channels (11-14) for the mixed gas flow or for the coolant, respectively,  
20
- a separating plate (16) without any opening for the inlet channel (11a) for the mixed gas flow is arranged between two successive stages (I, II),
- and separate apparatuses (10, 18) are provided for supplying oxidizing gas to the first and at least  
25 one other stage (I, II),

30 characterized  
in that the apparatus for supplying the oxidizing gas (18, 20, 21, 22) to the next stage (n+1, II) leads into the outlet channel (12a) for the mixed gas flow from the preceding stage (n, I).

35 2. Apparatus according to Claim 1,  
characterized

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in that three stages (III) are provided, in that all three stages (I-III) are in the form of a common plate stack, in that a further separating plate (16) without any opening (3) for the inlet channel (12b) for the mixed  
5 gas flow for the second stage (II) is arranged between the second and the third stage (II, III), in that a further apparatus (20) is provided for supplying the oxidizing gas to the third stage (III), and in that the apparatus for supplying the oxidizing gas (18) to the  
10 third stage (III) leads into the outlet channel (11b) for the mixed gas flow from the second stage (III).

3. Apparatus according to Claim 1,  
characterized

in that the apparatus for supplying the oxidizing gas  
15 (18) is in the form of a tubular probe which is passed through the end plate (15) or through the end plate (17) and the inlet channel (12b) for the second stage (II), into the outlet channel (12) from the first stage (I), and ends there.

20 4. Apparatus according to Claim 2,  
characterized

in that the apparatus for supplying the oxidizing gas (18) is in the form of a tubular probe which is passed through the end plate (15) or through the end plate (17),  
25 the outlet channel (12c) from the third stage (III), the separating plate (16) between the second and the third stage (II, III) and the inlet channel (12b) of the second stage (II), enter the outlet channel (12a) from the first stage (I), and ends there.

30 5. Apparatus according to Claim 2,  
characterized

in that the apparatus for supplying the oxidizing gas (20) is in the form of a tubular probe which is passed through the end plate (17) and, in the opposite direction  
35 to the mixed gas flow, through the inlet channel (11c) of the third stage (III), into the outlet channel (11b) from the second stage (II), and ends there.

6. Apparatus according to Claim 2,  
characterized  
in that the apparatus for supplying the oxidizing gas  
(20) is in the form of a tubular probe, which is passed  
5 through the supply channel (11a) of the first stage (I)  
and through the separating plate (16) between the first  
and the second stage (I, II), into the outlet channel  
(11b) from the second stage (II), and ends there.

7. Apparatus according to Claim 1,  
10 characterized  
in that the apparatus for supplying the oxidizing gas  
(18, 20, 21) is in the form of a tubular probe which has  
outlet openings (21.1, 21.2) whose geometric positions  
correspond to the reaction and/or cooling chambers (4, 5)  
15 of the preceding stage (n-1).

8. Apparatus according to Claim 7,  
characterized  
in that a plurality of outlet openings (21.1, 21.2) are  
arranged radially around the probe (21).

20 9. Apparatus according to Claim 7,  
characterized  
in that a plurality of outlet openings (21.1, 21.2) face  
the outlet channels (12a).

10. Apparatus according to Claim 1,  
25 characterized  
in that the apparatus for supplying the oxidizing gas is  
in the form of a tubular, coaxial probe (24), wherein an  
inner tube (22.1) at the end facing the preceding stage  
(n) has outlet openings (23) into an outer tube (22.2),  
30 and, on the side facing the next stage (n+1), the outer  
tube (22.2) has outlet openings (24) into the inlet  
channel of the next stage (n).

11. Apparatus according to Claim 1,  
characterized  
35 in that the apparatus for supplying the oxidizing gas is  
in the form of a tubular probe (5), wherein an inner tube  
(25.1) has one or more outlet openings (25.4) into a

deflection device (25.2) at the end facing the following stage (n+1) and, on the side facing the preceding stage (n), the deflection device (25.2) has one or more outlet openings (25.3) into the outlet channel (12a) from the 5 preceding stage (n) or the inlet channel (12b) of the next stage (n+1).

12. Apparatus according to one of the preceding claims,  
characterized

10 in that the oxidizing gas is supplied into the area between the outlet channel (12a) of the preceding stage (n) and the inlet channel (12b) of the following stage (n+1), in which area gas flows from the reaction chambers (4) are combined to form an overall volume flow.

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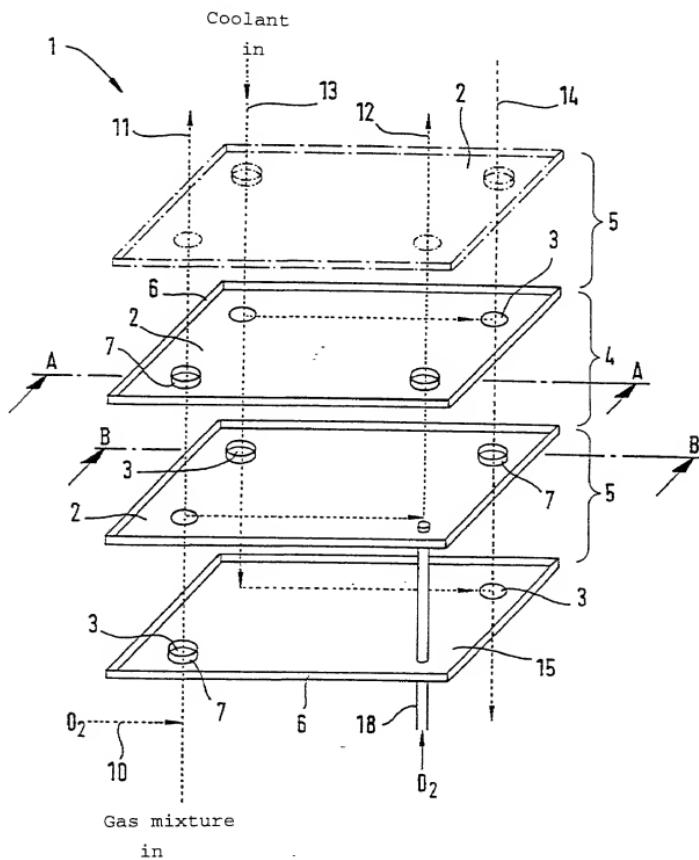
Abstract

The invention relates to an apparatus, in the form of plates, for selective catalytic oxidation of carbon monoxide contained in a hydrogen-rich mixed gas flow, wherein a reaction chamber filled with a catalyst or a cooling chamber through which a coolant flows is in each case formed between successive plates, and having openings to form inlet and outlet channels, respectively, for the mixed gas flow and the coolant, respectively, wherein the reaction chambers and cooling chambers, respectively, are each connected, for flow purposes, exclusively to the inlet and outlet channels for the mixed gas flow and for the coolant, respectively. According to the invention, the oxidizing gas is supplied directly into the inlet channel for the mixed gas flow. In the case of two-stage apparatuses, a plate without an opening for the inlet channel for the mixed gas flow is arranged between the first and the second stage, and the apparatus for supplying the oxidizing gas to the second stage is passed into the outlet channel for the mixed gas flow from the first stage.

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Fig.1



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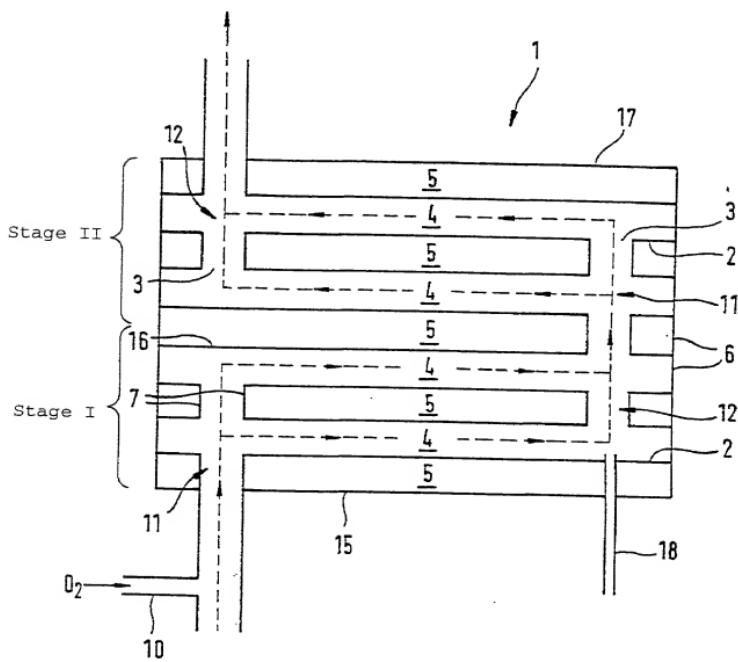


Fig. 2

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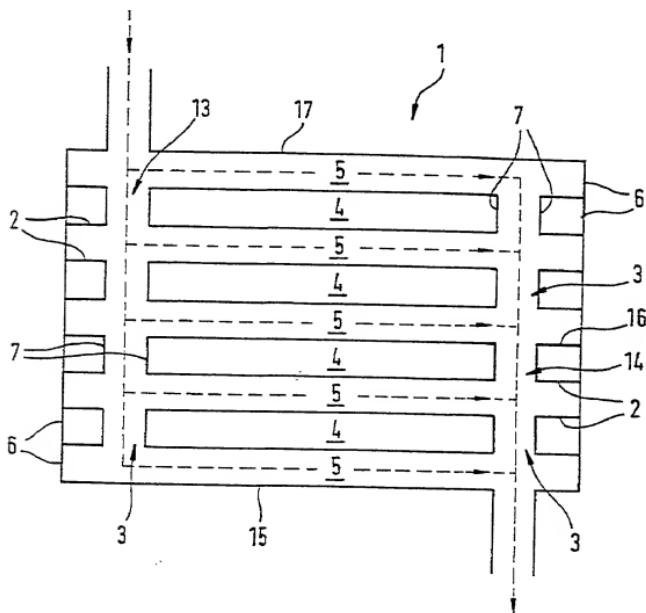


Fig. 3

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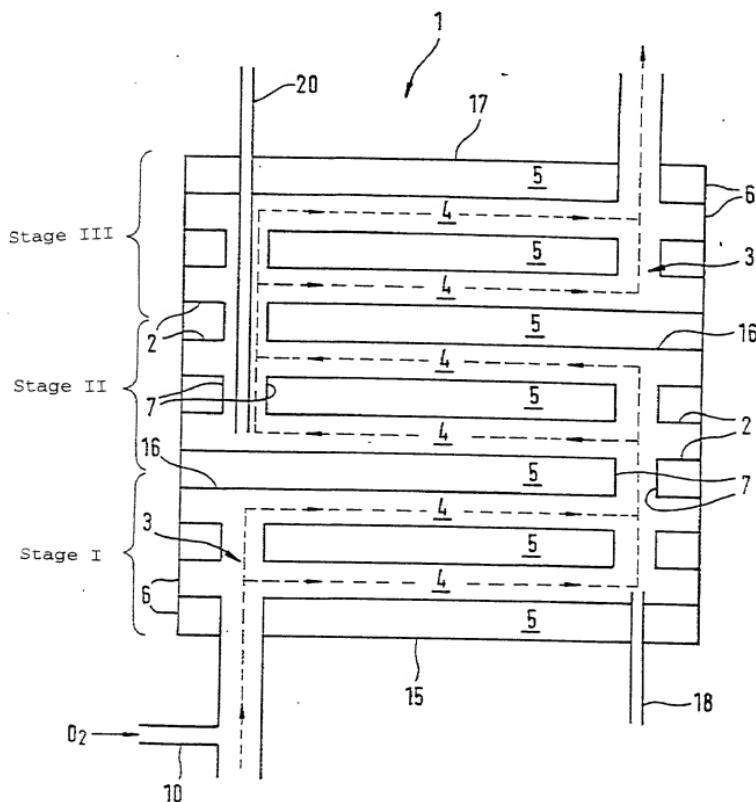


Fig. 4

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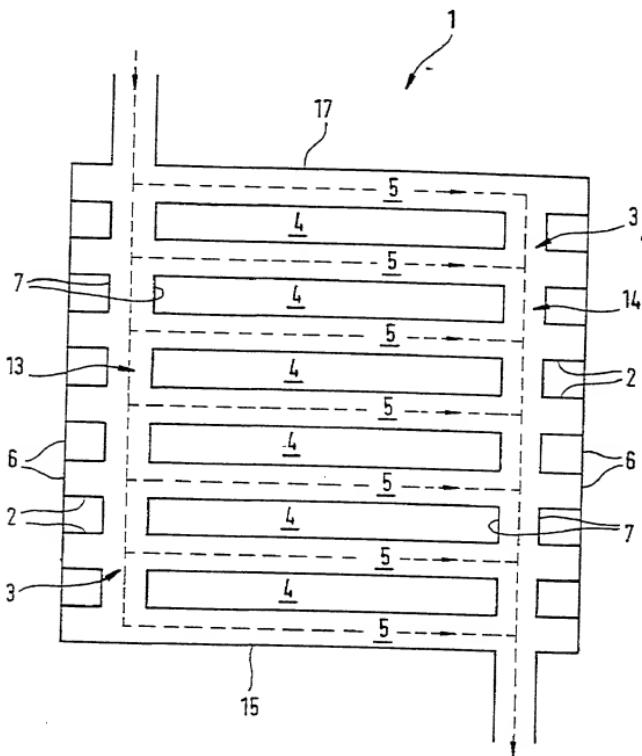


Fig. 5

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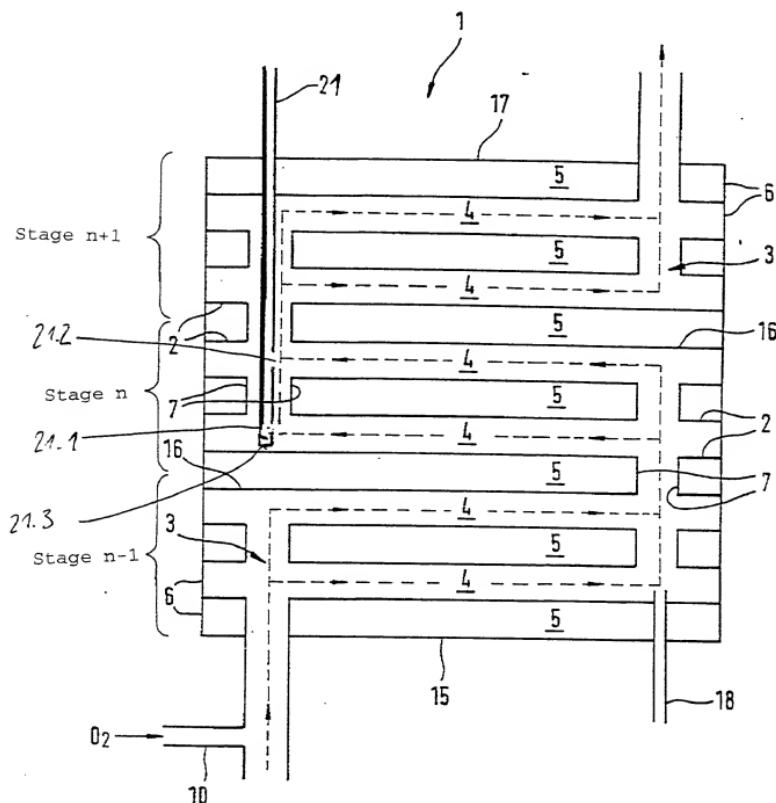


Fig. 6

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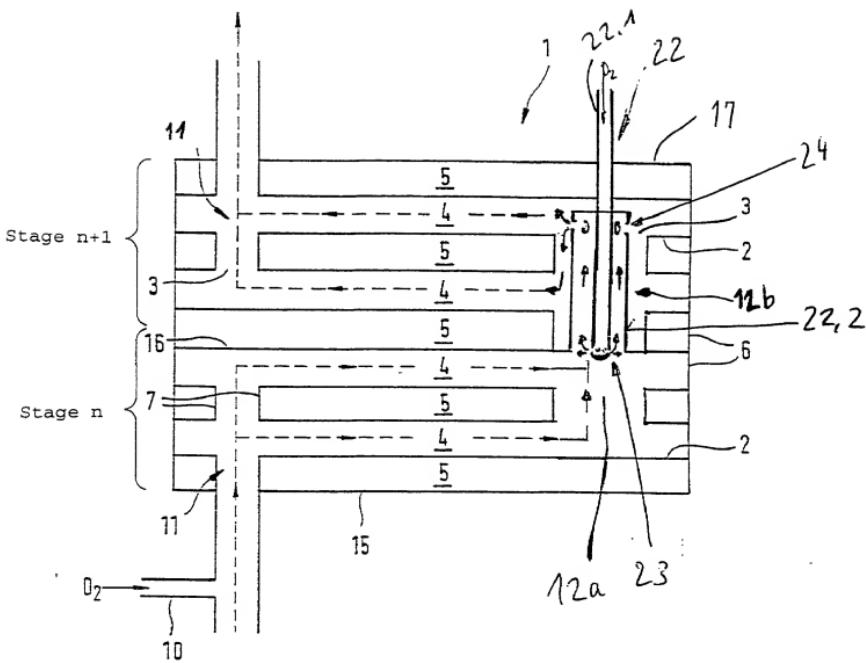


Fig. 7

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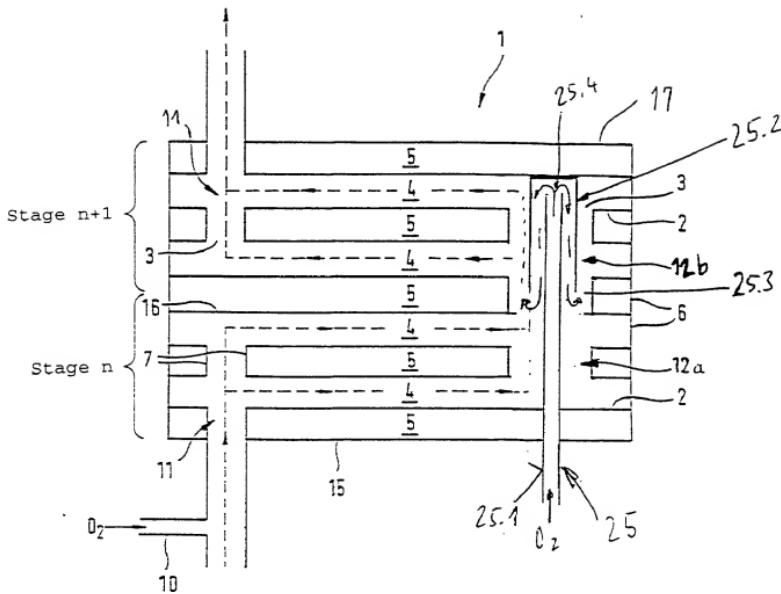


Fig. 8

## DECLARATION AND POWER OF ATTORNEY - PATENT APPLICATION

As a below named inventor, I hereby declare that my citizenship, postal address and residence are as stated below; that I verily believe I am the original, first and sole inventor (if only one inventor is named below) or a joint inventor (if plural inventors are named below) of the invention entitled:

## APPARATUS FOR SELECTIVE CATALYTIC OXIDATION OF CARBON MONOXIDE

the specification of which

       is attached hereto, or  
X was filed on December 3, 1998 as Application No. PCT/EP98/07868 and was amended on November 11, 1999.

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above. I acknowledge the duty to disclose all information known to be material to patentability as defined in 37 CFR §1.56. I hereby claim foreign priority benefits under Title 35, United States Code §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)	Priority Claimed		
<u>197 53 720.0</u> (Number)	<u>Germany</u> (Country)	<u>4 December 1997</u> (Day/Month/Year)	<u>Yes</u>

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose all information known to be material to patentability as defined in 37 CFR §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

<u>      </u> (Application Serial No.)	<u>      </u> (Filing Date)	<u>      </u> (Status)
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I hereby appoint as principal attorneys Herbert I. Cantor, Reg. No. 24,392; James F. McKeown, Reg. No. 25,406; Donald D. Evenson, Reg. No. 26,160; Joseph D. Evans, Reg. No. 26,269; Gary R. Edwards, Reg. No. 31,824; and Jeffrey D. Sanok, Reg. No. 32,169, to prosecute and transact all business in the Patent and Trademark Office connected with this application and any related United States and international applications. Please direct all communications to:

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under §1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

## DECLARATION AND POWER OF ATTORNEY

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Attorney Docket No. 225/48888

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2-cv

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